Direct comparison between two cold-atom-based optical frequency standards using a femtosecond-laser comb

K. R. Vogel, S. A. Diddams, C. W. Oates, E. A. Curtis, R. J. Rafac,
J. C. Bergquist, R. W. Fox, W. D. Lee, and L. Hollberg

Time and Frequency Division, National Institute of Standards and Technology, Boulder CO 80305-3328 USA

Using a fiber-broadened, femtosecond-laser frequency comb, the 76 THz interval between two laser-cooled optical frequency standards is measured with a statistical uncertainty of 2×10^{-13} in 5 s, the best short-term instability thus far reported for an optical frequency measurement. One standard is based on the calcium intercombination line at 657 nm, and the other on the mercury ion electric-quadrupole transition at 282 nm. By linking this measurement to the known Ca frequency, we report a new frequency value for the Hg^+ clock transition with an improvement in accuracy of $\sim 10^5$ over its best previous measurement.

Present state-of-the-art optical frequency standards based on cold atoms and ions exhibit excellent frequency stability and have the potential for achieving high reproducibility and accuracy. Such frequency references should find application in precise tests of fundamental physics and in next-generation atomic clocks. Incorporating these optical-based standards into a clockwork, however, has proven troublesome since their large frequencies (>300 THz) could not be conveniently converted to countable microwave signals. Several optical-frequency measurements have been made using harmonic chains to multiply up the frequency of the 9.2 GHz cesium microwave standard [1–4], but these chains are complex and require significant resources to operate. A paradigm-changing simplification was pioneered by Udem et al.[5] when they used the wide frequency-domain comb output of a femtosecond (fs) mode-locked laser to measure the absolute frequency of the Cs D_1 line and the hydrogen 1s–2s transition [6], the most accurate measurement to date of an optical frequency. A further refinement by Diddams et al. [7] expanded the available comb spectrum to an optical octave by broadening the fs-laser output in a microstructure fiber, leading to a direct connection between microwave and optical frequencies. A recent comparison of two independent, fiber-broadened fs-combs that measured the same frequency interval verified that a precision and reproducibility of $<5.1 \times 10^{-16}$ can be attained with this measurement method [8].

In this Letter we report a high-precision comparison between two very promising cold-atom optical frequency standards using a fs-laser frequency comb. One standard is based on a 2 mK collection of $\sim 10^7$ neutral ^{40}Ca atoms, and the other probes a single $^{199}Hg^+$ ion that is laser-cooled to near the Doppler limit. At 657 nm, a continuous-wave (cw), frequency-stabilized diode laser is locked to the central Ramsey-Bordé fringe obtained by four-pulse excitation of the Ca $^1S_0 - ^3P_1$ intercombination transition ($\nu_{Ca} = 456$ THz, $\Delta\nu = 400$ Hz) [9]. This system has demonstrated a frequency instability of $4 \times 10^{-15} \tau^{-1/2}$ (τ is the averag-
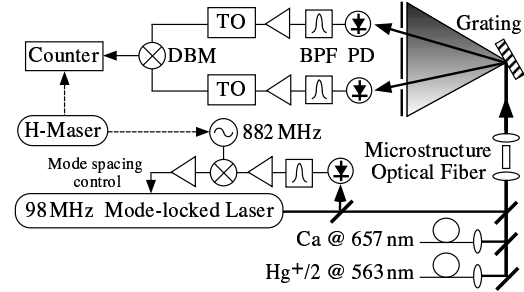

FIG. 1: Block diagram of frequency measurement. DBM: doubly-balanced mixer; PD: photodiode; BPF: bandpass filter; TO: tracking oscillator.

ing time) when probing sub-kilohertz fringe linewidths [10]. For the present measurements the Ca spectrometer was operated with 2.9 kHz linewidths, which gave an estimated short-term instability of $<2 \times 10^{-14} \tau^{-1/2}$. The oscillator in the Hg^+ standard is a frequency-narrowed cw dye laser at 563 nm that has a linewidth of about 0.16 Hz for a 20 s integration time [11]. This light is frequency-doubled to 282 nm to interrogate the $^2S_{1/2} - ^2D_{5/2}$ electric-quadrupole transition ($\nu_{Hg^+} = 1065$ THz, $\Delta\nu = 1.7$ Hz) of a Hg^+ ion that is confined in a linear, cryogenic, radio-frequency (rf) ion-trap. Rabi linewidths as narrow as 6.7 Hz at 282 nm have been observed with this system [12]. For these measurements the laser was stabilized to the Hg^+ ion with a linewidth of 40 Hz at 282 nm, and the instability under these conditions is estimated to be $<3 \times 10^{-15} \tau^{-1/2}$.

The fs-comb frequency measurement system, shown in Fig. 1, measures the 76 THz interval between the 563 nm and 657 nm light, which is transported from each stabilized laser by optical fiber. No attempt was made to actively cancel the fiber-added noise [11, 13], which we have measured to average down as $<4.4 \times 10^{-14} \tau^{-1/2}$ for these optical frequencies. The frequency comb is produced by a Kerr-lens, mode-locked Ti:sapphire laser that has a bandwidth of 42 nm

(FWHM) centered at 810 nm and operates with a repetition rate of ≈98 MHz. To control the frequency spacing between comb modes, the ninth harmonic of the repetition rate is detected by a high-speed photodiode with a signal-to-noise ratio (S/N) of >70 dB in a 100 kHz bandwidth (BW), and then mixed with an 882 MHz reference signal from a frequency synthesizer. The repetition rate is phase-locked to this reference frequency by using the phase-dependent mixer output to control the horizontal tilt of the high-reflector mirror, which is situated after a dispersion-compensating prism-pair [5, 6]. The internal clock of the synthesizer is phase-locked to a hydrogen maser [H-maser, $\sigma_y(\tau) \approx 2 \times 10^{-13}\,\tau^{-1/2}$], whose frequency is known with an uncertainty of $\pm 4 \times 10^{-15}$ by comparison to the NIST Cs primary standard.

To extend the comb bandwidth, the fs-pulses are launched into a 5 cm-long microstructure optical fiber that has a core diameter of ≈1.7μm and zero group-velocity dispersion near 770 nm. Self-phase modulation and other nonlinearities in the fiber produce an output spectrum from about 500 to 1100 nm [14]. The cw light from both stabilized lasers is also coupled into the microstructure fiber to ensure good spatial mode overlap with the fs-laser light. The fiber output is dispersed by an optical grating and imaged onto slits so that only a few modes in the vicinity of the cw light frequencies are selected. Photodiodes after each slit detect the rf heterodyne beatnotes (δ_1, δ_2) between the cw light and a comb mode with a S/N of ~25 dB in a 100 kHz BW, with the background limited by the shot-noise of the cw light power. For accurate frequency counting, tracking oscillators are phase-locked to the beatnotes to provide regenerated signals with >50 dB S/N in a 100 kHz BW. Since the comb-mode spacing ν_{rep} is locked to the H-maser, we can remove the fs-laser frequency jitter (~10 MHz) common to both beatnotes by mixing together the correlated signals from the two tracking oscillators [15]. The mixer output is either a stable sum or difference signal $\delta = \delta_1 \pm \delta_2$, which is counted to yield the frequency interval $\Delta\nu = \frac{1}{2}\nu_{Hg^+} - \nu_{Ca} = N\nu_{rep} \pm \delta$. The integer number N and the sign choice for δ were determined unambiguously by comparing to our previous ±10 MHz measurement of this frequency difference [16], and verified by making measurements for different repetition rates.

The inset in Fig. 2 shows a typical time record of the frequency fluctuations of δ, counted with a 5 s gate time. Points that exhibit obvious cycle slip errors, predominantly due to a tracking oscillator losing lock, are eliminated in the data sorting. The Allan deviation, shown in Fig. 2, indicates that the measurement precision averages down as $(34\,\text{Hz})\,\tau^{-1/2}$ for the duration of the measurement. Three frequency sources (ν_{Ca}, ν_{Hg^+}, ν_{rep}) contribute to the short-term instability of this measurement, and from this data alone we cannot attribute the noise unambiguously to a specific source. Nonetheless, by assuming that all of the noise comes solely from a given source, we can place an upper limit on its short-term Allan deviation. From this data
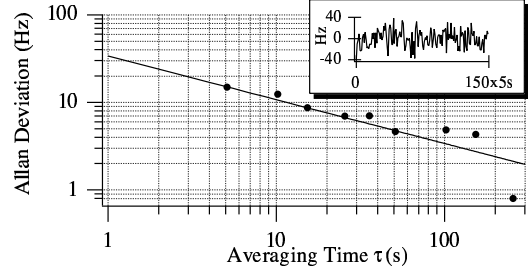

FIG. 2: Allan deviation of typical measurement record. The line is $(34\,\text{Hz})\,\tau^{-1/2}$, and the inset shows the corresponding time record. From this data, we place upper limits of $\sigma_y(\tau) \leq 45$, 7.4, $6.4 \times 10^{-14}\,\tau^{-1/2}$ for the short-term instability of the microwave reference, ν_{Ca}, and ν_{Hg^+}, respectively.

we infer an upper limit of $\leq 7 \times 10^{-14}\,\tau^{-1/2}$ for the fractional frequency instability of the two optical standards, although each probably has a significantly better stability [10–12]. Combining the best estimates for the (normalized) instabilities of the microwave and two optical references, as well as for the optical fiber delivery, we arrive at a calculated instability of $3.5 \times 10^{-13}\,\tau^{-1/2}$ for the 76 THz interval, in good agreement with the measured value of $4.5 \times 10^{-13}\,\tau^{-1/2}$. There are likely additional degradations of the stability due to noise in the microwave detection of the repetition rate and/or in the synthesizer electronics that multiply up the frequency of the H-maser.

The results of running the fs-comb measurement system on four separate days over a six week period are plotted in Fig. 3 as the frequency offset from the weighted mean, which is $\Delta\nu = 76\,374\,564\,455\,467\,(30)\,\text{Hz}$. Each of these points represents the weighted mean of the data runs on an individual day, corrected for the second-order Zeeman shifts for both Ca and Hg$^+$, which are determined to an uncertainty of $<10^{-14}$ for each day. The Ca system contributes an estimated uncertainty of 40 Hz to the total measurement uncertainty, while the Hg$^+$ reference is known to <10 Hz (at 563 nm), limited by the current measurement of the electric-quadrupole shift [12]. The largest systematic error (~30 Hz) stems from uncertainty in our knowledge of the angular overlap of the counterpropagating probe beams in the Ca spectrometer, which leads to a residual first-order Doppler shift when the cold Ca ensemble has a transverse drift velocity [2]. It is noteworthy that the uncertainties for the data of May 5 and 25 are dominated by systematic effects. The run-to-run measurements on those days are consistent to ≈10 Hz, as illustrated for the May 25 data in the Fig. 3 inset, which shows only the statistical uncertainty for each run. Given the relatively small statistical contribution (~2 Hz on a given day) to the total uncertainty, we believe that fs-laser-based frequency metrology gives us, for the first time, a practical tool for evaluating systematic shifts at an inaccuracy approaching 10^{-15} for these high-accuracy optical standards.
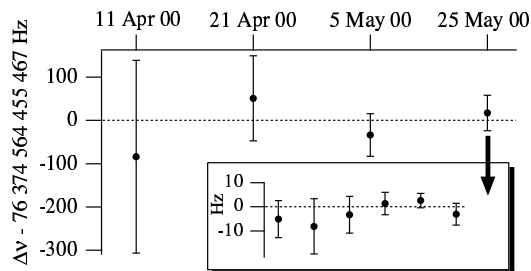
FIG. 3: Frequency deviation in Hz of $\Delta\nu = \frac{1}{2}\nu_{Hg^+} - \nu_{Ca}$. Each data point is the weighted mean of an individual day's data runs. Inset shows the data runs for May 25 collected over a 50 min period, with each run averaging 230 s. Error bars in the inset reflect only statistical uncertainties.

The uncertainties given for the two optical references are estimated for conditions during these experiments only, and no serious attempt was made to minimize systematic effects. Nevertheless, it is encouraging that our results for the frequency difference have a standard deviation of the mean of 60 Hz over a six week period. This consistency shows that all three components, the frequency-measurement system and the two optical standards, are reproducible at this level. Thus, we offer an improved value for the Hg^+ clock transition frequency by summing our measured $Hg^+/2-Ca$ interval with the absolute frequency of the Ca 657 nm clock transition measured by the PTB multiplication chain [2]. We obtain $\nu_{Hg^+} = 2 \times 532\,360\,804\,949\,597\,(124)$ Hz, where the uncertainty is dominated by the 120 Hz uncertainty in the Ca measurement. This is an 80,000-fold improvement over the best previous frequency measurement of the 282 nm clock transition [16].

Work is currently underway to self-reference the frequency offset of our fs-comb [17], and we anticipate confirmation of the Hg^+ and Ca frequencies with a direct rf-to-optical measurement. In fact, by locking a mode of a self-referenced comb to one of the optical standards, an all-optical connection between Hg^+ and Ca could be achieved that eliminates any dependence on the H-maser microwave reference. This interval measurement should then average down at the stability of the optical standards alone, allowing for more precise determination of systematic shifts. The capability to intercompare three high-performance frequency standards (Hg^+, Ca, and Cs) has powerful advantages, such as determining absolute frequency stabilities and testing the fidelity of fs-comb measurements. Indeed, an optical clock is realized by the repetition-rate output of an all-optically-referenced comb, and microwave sources with frequency instabilities near $10^{-15}\tau^{-1/2}$ should be obtainable provided that the repetition-rate signal can be extracted with a suitably high signal-to-noise ratio.

REFERENCES

[1] D. A. Jennings, C. R. Pollock, F. R. Petersen, R. E. Drullinger, K. M. Evenson, J. S. Wells, J. L. Hall, and H. P. Layer, Opt. Lett. **8**, 136 (1983); C. R. Pollock, D. A. Jennings, F. R. Petersen, J. S. Wells, R. E. Drullinger, E. C. Beaty, and K. M. Evenson, Opt. Lett. **8**, 133 (1983).

[2] H. Schnatz, B. Lipphardt, J. Helmcke, F. Riehle, and G. Zinner, Phys. Rev. Lett. **76**, 18 (1996); F. Riehle, H. Schnatz, B. Lipphardt, G. Zinner, T. Trebst, T. Binnewies, G. Wilpers, and J. Helmcke, in *Proceedings of the 1999 Joint Meeting of the EFTF - IEEE IFCS*, eds. R. J. Besson, D. B. Sullivan, (1999), pp. 700–705.

[3] D. Touahri, O. Acef, A. Clairon, J.-J. Zondy, R. Felder, L. Hilico, B. de Beauvoir, F. Biraben, and F. Nez, Opt. Commun. **133**, 471 (1997).

[4] J. E. Bernard, A. A. Madej, L. Marmet, B. G. Whitford, K. J. Siemsen, and S. Cundy, Phys. Rev. Lett. **82**, 3228 (1999).

[5] Th. Udem, J. Reichert, R. Holzwarth, and T. W. Hänsch, Opt. Lett. **24**, 881 (1999); Phys. Rev. Lett. **82**, 3568 (1999).

[6] M. Niering, R. Holzwarth, J. Reichert, P. Pokasov, Th. Udem, M. Weitz, T.W. Hänsch, P. Lemonde, G. Santarelli, M. Abgrall, P. Laurent, C. Salomon, and A. Clairon, Phys. Rev. Lett. **84**, 5496 (2000).

[7] S. A. Diddams, D. J. Jones, J. Ye, S. T. Cundiff, J. L. Hall, J. K. Ranka, R. S. Windeler, R. Holzwarth, Th. Udem, and T. W. Hänsch, Phys. Rev. Lett. **84**, 5102 (2000).

[8] R. Holzwarth, Th. Udem, T. W. Hänsch, J. C. Knight, W. J. Wadsworth, and P. St. Russell, accepted for publication in Phys. Rev. Lett.

[9] C. W. Oates, F. Bondu, R. W. Fox, and L. Hollberg, Eur. Phys. J. D. **7**, 449 (1999).

[10] C. W. Oates, E. A. Curtis, and L. Hollberg, accepted for publication in Opt. Lett.

[11] B. C. Young, R. J. Rafac, J. A. Beall, F. C. Cruz, W. M. Itano, D. J. Wineland, and J. C. Bergquist, in *Proceedings of XIV International Conference on Laser Spectroscopy*, (World Scientific, 1999), pp. 61–70.

[12] R. J. Rafac, B. C. Young, J. A. Beall, W. M. Itano, D. J. Wineland, and J. C. Bergquist, accepted for publication in Phys. Rev. Lett.

[13] L-S. Ma, P. Jungner, J. Ye, and J. L. Hall, Opt. Lett. **19**, 1777 (1994).

[14] J. K. Ranka, R. S. Windeler, and A. Stentz, Opt. Lett. **25**, 25 (2000).

[15] S. A. Diddams, D. J. Jones, L-S. Ma, S. T. Cundiff, and J. L. Hall, Opt. Lett. **25**, 186 (2000).

[16] B. Frech, J. S. Wells, C. W. Oates, J. Mitchell, Y-P. Lan, T. Kurosu, L. Zink, L. Hollberg, T. Zibrova, B. C. Young, and J. C. Bergquist, IEEE Trans. Ultr. Ferr. Freq. Cont. **47**, 513 (2000).

[17] D. J. Jones, S. A. Diddams, J. K. Ranka, A. Stentz, R. S. Windeler, J. L. Hall, and S. T. Cundiff, Science **288**, 635 (2000).